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Article

Impact of Rain Precipitation on Urban Atmospheric Particle Matter Measured at Three Locations in France between 2013 and 2019

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Abstract: As atmospheric particle matter (PM) pollution has been proven to be a public health risk, we investigated how PM concentrations of various particle diameters may be impacted by precipitation. Repeated measures over time of urban PM concentrations for particles of 0.2–50 µm in diameter were compared with precipitation data from Météo-France weather stations in Paris, Angers and Palaiseau from 2013 to 2019. A significant negative correlation, using Kendall's rank correlation, was found between the amount of precipitation and concentrations of particles >3 µm. Distribution comparative analysis (Dunn's test) of 154 events of 1 mm or more of rain demonstrated a decrease in concentrations for particles from 10 to 50 µm in diameter. Additionally, granulometric analysis of a typical heavy rain event showed a 10-fold decrease in concentrations of particles 10 to 30 µm in diameter one hour after rain compared with one hour before. We were able to show that measured concentrations of particles between 10 and 50 µm in diameter diminish when it rains, with a lasting effect of approximately 10–15 h.

Keywords: particle matter; precipitation; rain; LOAC; Paris; Angers; air pollution; particulate matter

1. Introduction

Public health organizations alert us to the major growing health risk of air pollution. In March 2014, a study by the World Health Organization (WHO) indicated that in 2012, the death of 7 million people worldwide was due to air pollution [1], a figure destined to increase as shown in a 2018 study by the Proceedings of the National Academy of Sciences of the United States of America (PNAS) estimating the figure to be closer to 9 million per year [2]. These studies looked at µg/m³, whereas the current study uses count/cm³.

Among the air pollutants, particle matter (PM) has harmful effects on several organs, the most documented of them being the respiratory system, the cardiovascular system and the central nervous system [3]. The extent and type of harm depends on the particle size. The finer a particle is, the deeper it can penetrate into the human body via the airways and the lungs. The finest particles can reach other organs through blood circulation [4]. Particles suspended in the air can be emitted by human activities, such as transportation, traffic, heating, construction sites, industrial activities and agriculture. They can also be of natural origin (volcanic eruption, forest fire, wind erosion, desert dust) [5]. The lifetime of particles in the air can vary according to weather conditions and size [6]. Further, some particles can remain suspended and be transported over long distances [7].

During anticyclonic conditions, PM concentrations can strongly increase because of low winds and local sources of pollution [8]. Likewise, pollution levels are expected to be

lower during windy and rainy conditions that can disperse, wash and clean the ambient air. Thus, meteorological conditions (e.g., rain, atmospheric pressure, wind speed and direction) are key parameters to be considered when studying urban PM pollution and its variability. If the washing effects of rain on PM content can be determined across particle sizes, this could help evaluate and predict local air quality variability and citizens' exposure to pollution during various weather conditions.

A previous study explored the mean seasonal behavior of ultrafine particles (15–660 nm) and its correlation with precipitation [9]. Another study examined the behavior of fine and coarse particles under diverse meteorological conditions [10], showing that 1.6 mm of rainfall significantly reduced PM_{2.5} levels. These studies concerned mass concentrations of particles of all diameters smaller than 2.5 micrometers that could have been dominated by the heaviest PM_{2.5} particles. Since the smallest particles are the most dangerous, it is worthwhile to study multiple size ranges of small particle concentrations.

In this paper, we specifically examine the correlation between precipitation and urban pollution PM number concentrations in the ambient air across discrete size ranges from 0.2 to 50 μm in diameter at three urban sites in metropolitan France. Measurements were obtained by the Light Optical Aerosol Particles Counter (LOAC) instrument, which counts particles by specific size range per cubic centimeter. These particle counts by size range are more precise than mass concentration levels when analyzing the impact of rain on particles [11].

2. Materials and Methods

2.1. Particle Matter Measurement, the LOAC

The LOAC is an instrument developed for ground- and balloon-based PM measurements [11,12]. The air sampling is performed through a total suspended particulate (TSP) inlet vertically oriented and having a diameter cut-off at about 100 μm . Such a system prevents collection of rain droplets larger than 100 μm . Particles are drawn up to the optical chamber through an isostatic tube by a small pump and cross a laser beam working at 650 nm. The scattered light is recorded by two photodiodes at scattering angles of $\sim 15^\circ$ and $\sim 65^\circ$, and photons travel directly to the photodiodes through pipes without a lens. A total of 19 size classes are defined in the particle diameter range between 0.2 and $\sim 50 \mu\text{m}$.

The LOAC provides measurements every 10 s. To reduce the dispersion of the measurements, they are averaged over 10–15 min. For such integration time, the measurement uncertainty is about $\pm 20\%$ for concentrations higher than 10 particles per cubic centimeter and up to $\pm 60\%$ for concentrations lower than 1 particle per cubic centimeter. The lower detection limit is about 0.1 particles per cubic centimeter. The measurement uncertainty in size determination is of $\pm 0.025 \mu\text{m}$ for particles smaller than 0.6 μm , $\pm 5\%$ for particles in the 0.7–2 μm range and $\pm 10\%$ for particles greater than 2 μm .

The performance of the LOAC has been established over numerous sessions of inter-comparison with other instruments dedicated to the counting, the size distribution, the extinctions and mass concentrations of solid aerosols in the atmosphere [12]. These sessions have shown that the LOAC can be used for studies of urban aerosols. The measurements of PM concentrations for each of the ranges taken every 10–15 min were averaged over each hour for this study to match with the precipitation data.

2.2. Particle Matter Measurement Locations

Three LOAC stations measured PM concentrations continuously over several years with little downtime, in Paris, Palaiseau and Angers. The first one was installed on the basket of the touristic tethered balloon “Ballon de Paris Generali” (Figure 1) in the André Citroën park in southwestern Paris, France (48.8414° N, 2.2740° E). It has been operational since mid-2013 [13]. The balloon is not in flight during windy and rainy conditions; nevertheless, the measurements are conducted even when the balloon is on the ground. The second LOAC is at the SIRTa observatory (48.7180° N, 2.2073° E) located in a semi-urban area, in Palaiseau, 25 km south of Paris, France [13,14]. It has been operating since

mid-2015. The third one is on the gondola of the touristic balloon in the park “Terra Botanica” in northern Angers, France (47.4970° N, 0.5701° W). It was placed there in January 2017.

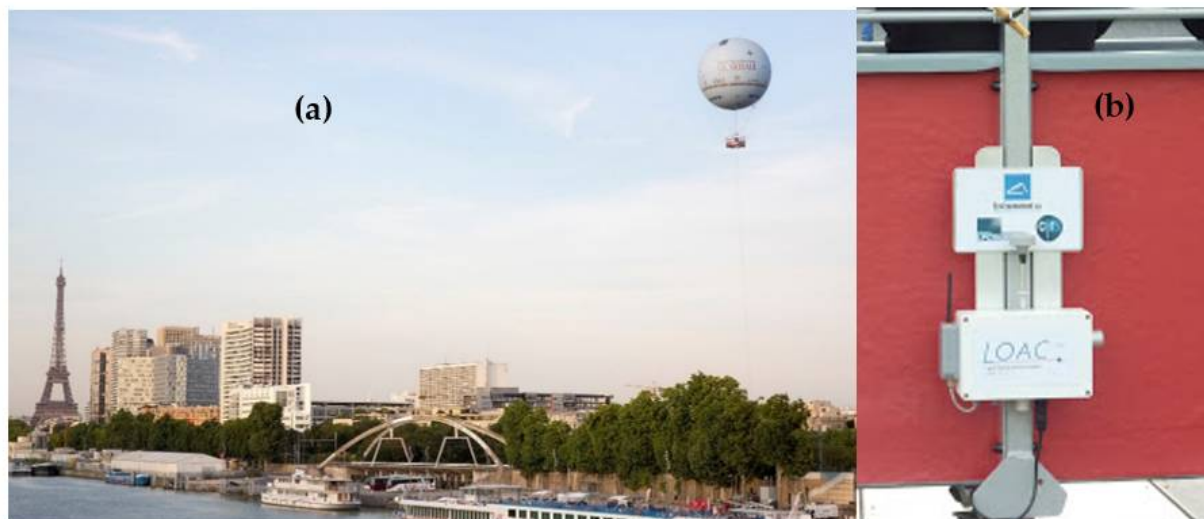


Figure 1. (a) Generali balloon above the André Citroën park in Paris; (b) the LOAC attached to the Generali balloon (We do own this image, it is free to use under the creative commons license: <https://creativecommons.org/licenses/by-sa/4.0/deed.en>).

2.3. Precipitation Data

Météo-France, the national weather service in France, provided us with full access to all the precipitation data between 2013 and 2019 that we used for this study as a teaching and research grant.

As measured by Météo-France, ‘precipitation’ in millimeters includes the melted liquid equivalent of snow and other frozen precipitation. However, snow represented only 1.7% of precipitation in Paris over the course of the study and less than 0.2% of the precipitation in Angers. Rain and drizzle falling to the ground before evaporation account for the vast majority of the precipitation in this study. The terms ‘rain’ and ‘precipitation’ are used interchangeably in this paper to refer to precipitation measurements.

Météo-France recorded all episodes of precipitation yielding at least 0.2 mm of rain in one hour. These measurements were taken in increments of 0.1 mm providing an accumulated total over the hour. Of the 85,232 h of LOAC particle matter measurements at the three locations, 7147 had precipitation (8.39%).

We were able to use hourly precipitation data collected from the three meteorological stations closest to the particle measurement locations (Figure 2) and in similar geographical conditions: Montsouris ($+48.8217$, $+2.3378$; 5 km from the Generali tethered balloon in the André-Citroën park), Angers-Beaucouzé ($+47.4782$, -0.6144 ; 4.1 km from the tethered balloon at Terra Botanica in Angers) and Villacoublay-Vélizy ($+48.7725$, $+2.2039$; 7.5 km from the SIRTa campus in Palaiseau).

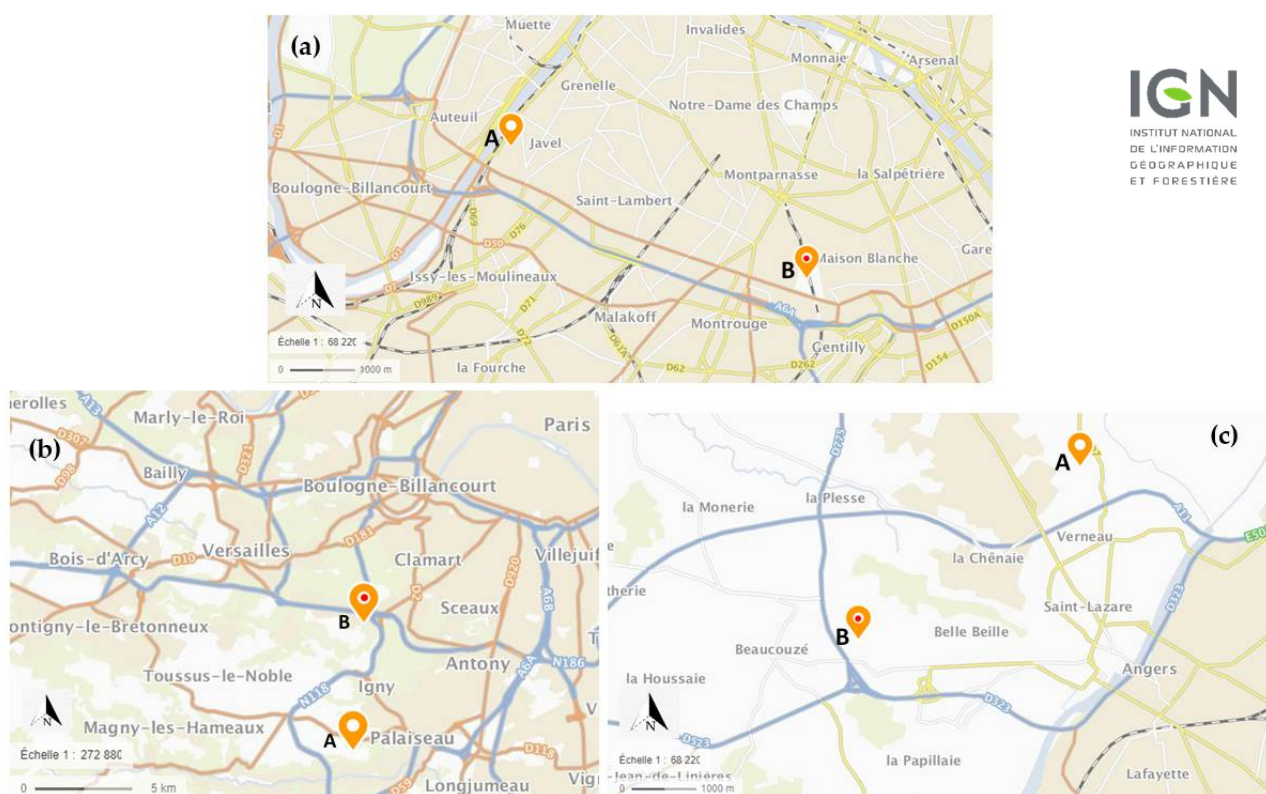


Figure 2. Maps showing the location of the LOAC (A) and the corresponding weather station (B). (a) In Paris, 5 km apart; (b) In Palaiseau, 7.5 km apart; (c) In Angers, 4.1 km apart (@ IGN 2020).

2.4. Statistical Methods

2.4.1. Descriptive Statistics and Correlations

To begin, all 85,232 time interval measurements for all years and for all three LOAC sites were analyzed. This established a baseline of concentration level distributions described using a histogram to provide frequency for each particle size range, allowing us to visualize the data and determine normal distribution. To reduce the natural dispersion of the measurements, the 19 particle sizes were grouped into six size ranges: 0.2–0.5 μm , 0.5–1.1 μm , 1.1–3 μm , 3–10 μm , 10–20 μm and 20–50 μm . These size ranges could be representative of the commonly used PM_{0.5}, PM₁–PM_{0.5}, PM_{2.5}–PM₁, PM₁₀–PM_{2.5}, PM₂₀–PM₁₀ and PM₅₀–PM₂₀ particles (PM meaning particle matter smaller than a given size).

Similarly, the total accumulated precipitation measurements for each hour were analyzed for the same years and for all three weather stations.

Further, a non-parametric correlation using Kendall's tau [15] was then used to analyze the relationships between precipitation and PM concentrations for each size range. This test was used because the PM and precipitation data were not normally distributed.

All statistics for this study were run with RStudio using version 3.6.2 of R.

2.4.2. Granulometric Analyses

In the second part of this study, a typical heavy rain event was selected to observe how PM concentrations varied in the hours following a single rain event. The episode of rain lasted five hours and yielded 10.6 mm of rain. No precipitation had been measured for the previous 56 h leading up to this rain event. Further, it did not rain during the 15 h following the event, permitting an analysis of the recuperation of PM concentrations. The episode of rain began on 8 June 2017 at 11:00 p.m. at the Paris-Montsouris weather station.

The granulometric analysis plots grain size against the percentage of the individual size fractions in a specific sample. Three temporal samples were chosen to compare the PM concentrations 1 h before the rain event began with PM concentrations in the hour following the rain event and with concentrations 15 h after the rain event. In a granulometric analysis, the PM concentrations for each size range are divided by the midpoint of the size range in order to provide a relative measure of concentrations. These results were then plotted on the Y-axis against the median particle size on the X-axis. A line was drawn through the resulting points for each 1 h period plotted on the log graph, allowing visual comparisons between periods across the spectrum of particle sizes. Enough data were available for each of the 19 size ranges to be used for these observational analyses.

2.4.3. Comparative Analyses of PM Concentration Distributions before, during and after Rain

In the final part of the study, PM concentrations for each particle size interval were analyzed from 5 h before rain to 20 h after rain events stopped. We chose to study up to 20 h after the rain events (four 5 h periods), and no more, to maintain a large sample size (154) of rain events meeting the criteria of not having been preceded by or followed by rain for at least 21 h. As with the descriptive statistics, the 19 particle sizes were grouped into six size ranges. The rain events studied are those that produced at least 1 mm of precipitation, as a previous study found that 1.6 mm of rainfall significantly affected PM_{2.5} levels [10]. A total of 1 mm of rain corresponds to 1 L of water over 1 square meter. Additionally, the rain events studied were not preceded by or followed by rain for at least 21 h in order to have enough time before and after rain to observe the evolution of PM concentrations. These criteria resulted in 154 rain events for analysis.

Six graphs, one for each particle size range, were made to show the evolution of median particle matter concentrations from 5 h before to 20 h after the rain events. Bars below and above the median were added to show the 40th and 60th percentiles. These percentiles were chosen to indicate the midrange of concentrations—lower and upper quartiles or deciles would not have demonstrated as much of a meaningful pattern due to the high variance across the rain events.

Dunn's test [16], which is a non-parametric comparative analysis of distributions, was then used to compare the PM concentrations for various time periods before and after rain events. PM concentrations for each of the six size ranges were grouped into six periods to simplify the analysis:

- 5–1 h before the rain events,
- The periods of rain (durations varied) rounded to the closest hour,
- 1–5 h after the rain events,
- 6–10 h after the rain events,
- 11–15 h after the rain events,
- 16–20 h after the rain events.

The difference between distributions was considered significant if the *p*-value was inferior to 0.025 (5% probability of statistical error) for this test.

3. Results

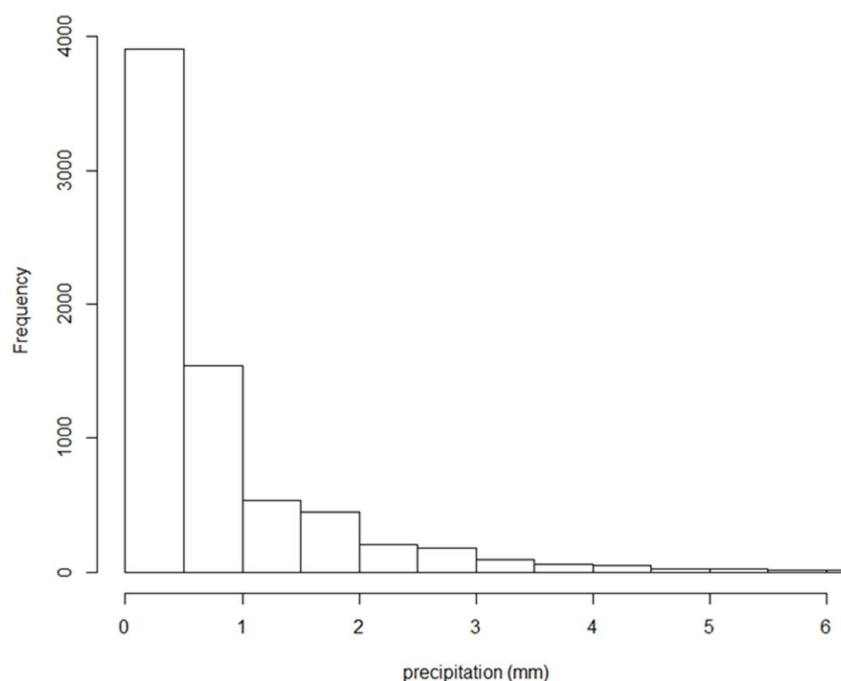
3.1. Descriptive Statistics of the Data and Correlation

3.1.1. Description of Precipitation Data

The total accumulated precipitation levels in millimeters were recorded hourly. The distribution of these levels is described in Table 1. The histogram allows us to visualize the data (Figure 3). We can see that the precipitation data are not normally distributed. This violates the assumptions of conventional correlation tests (Pearson) and comparison for correlated groups tests (ANOVA). Therefore, we must use non-parametric tests; in this case, we chose Kendall's rank correlation and Dunn's pairwise tests.

Table 1. Descriptive statistics of accumulated precipitation levels (mm) for each hour of precipitation in three locations between 2013 and 2019.

Descriptive Statistics	Values
Sample size	7147
Minimum considered	0.2
1st quartile	0.2
Median	0.4
3rd quartile	1.0
Maximum	49.2
Mean	0.91
Standard deviation	1.48

**Figure 3.** Histogram of precipitation indicating the number of hours of observations (frequency) of different amounts of precipitation (mm) in the three locations between 2013 and 2019.

3.1.2. Description of PM Distributions

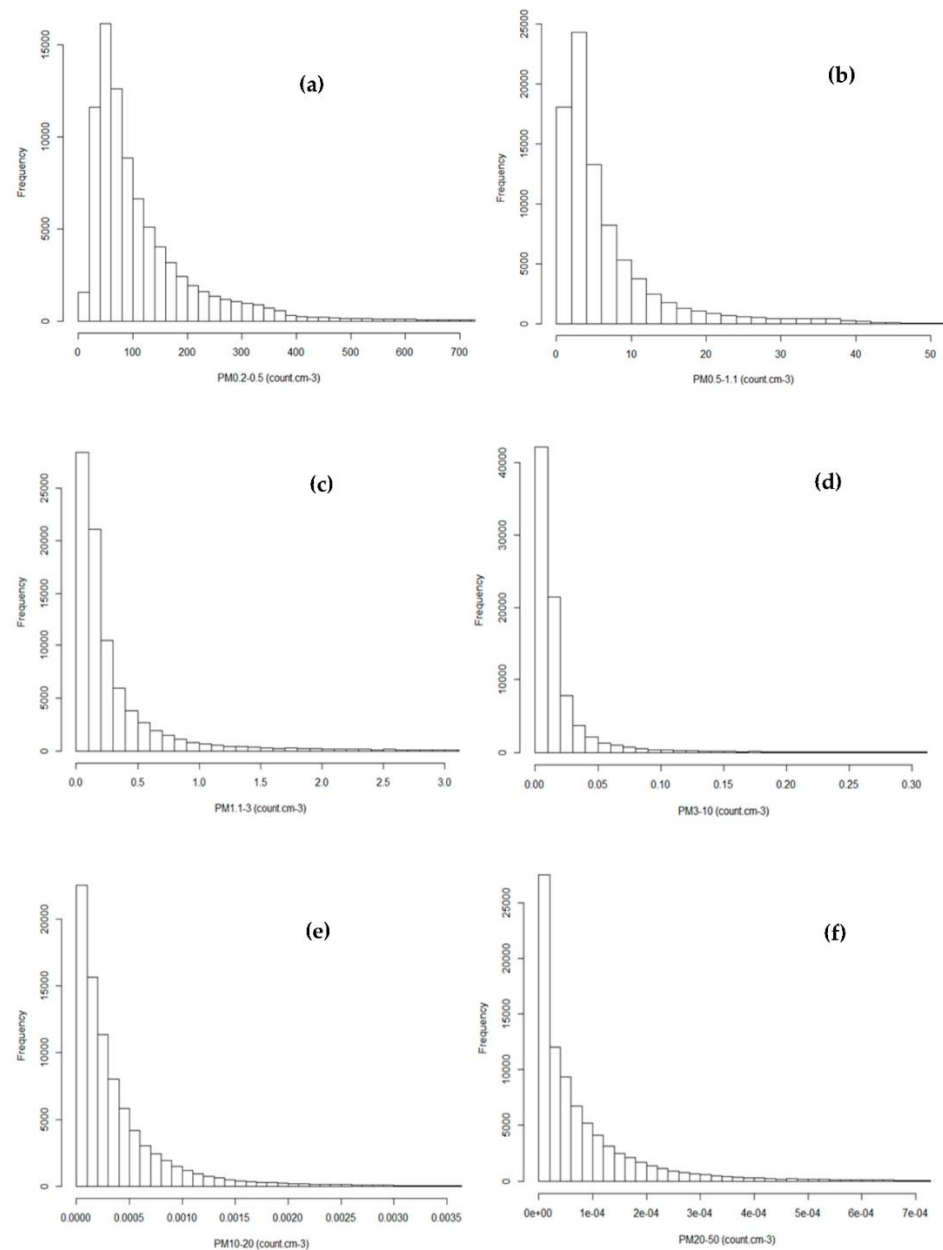
Hourly averages of concentrations for each of the six particle size ranges were recorded over six years at three locations:

- Angers, from 19 January 2017 to 31 December 2019;
- Palaiseau, from 23 March 2016 to 31 December 2019;
- Paris, from 24 March 2013 to 31 December 2019.

This resulted in approximately 85,000 measurements. A summary of standard descriptive statistics for each of the six size ranges is shown in Table 2. Distributions of PM data for each size interval are visualized using histograms (Figure 4). The PM data are not normally distributed. This violates the assumptions of conventional correlation tests (Pearson) and comparison for correlated groups tests (ANOVA). Therefore, we used non-parametric tests; in this case, we chose Kendall's rank correlation and Dunn's pairwise tests.

Table 2. Descriptive statistics of the PM concentrations in the three locations between 2013 and 2019.

	0.2–0.5 μm	0.5–1.1 μm	1.1–3 μm	3–10 μm	10–20 μm	20–50 μm
Sample size	85,232	85,232	85,232	85,232	85,232	85,232
Minimum	0.36	0	0	0	0	0
1st quartile	50	2.2	0.08	0.005	0.00009	0.00001
Median	81	4.0	0.16	0.010	0.0002	0.00005
3rd quartile	146	8.0	0.36	0.020	0.0005	0.0001
Maximum	10695	1010	111	123	51	24
Mean	128	7.49	0.65	0.25	0.033	0.007
Standard deviation	196	16.64	3.35	2.98	0.630	0.223

**Figure 4.** Histogram indicating the number of observations (frequency) of different PM concentrations (count per cm³) for particles of size ranges: (a) 0.2–0.5 μm ; (b) 0.5–1.1 μm ; (c) 1.1–3 μm ; (d) 3–10 μm ; (e) 10–20 μm and (f) 20–50 μm in three locations between 2013 and 2019.

3.2. Correlation between Precipitation and PM Concentrations

Correlation results between precipitation amounts and PM concentrations of the six size ranges are shown in Table 3. The PM and precipitation data are not normally distributed so a non-parametric test, Kendall's rank correlation [15], was used to analyze the relationships between precipitation and PM concentrations.

Table 3. Kendall's rank correlation results between precipitation and PM in three locations between 2013 and 2019.

	0.2–0.5 μm	0.5–1.1 μm	1.1–3 μm	3–10 μm	10–20 μm	20–50 μm
Tau	−0.004	−0.002	−0.003	−0.041	−0.050	−0.035
p-value	0.144	0.508	0.364	$2.2 \times 10^{-16} *$	$2.2 \times 10^{-16} *$	$2.2 \times 10^{-16} *$

* Indicates statistical significance.

The amount of precipitation accumulated over each of the 85,232 observed hours was compared with the average PM concentrations of those same hours. A total of 7147 of those hours had measurable precipitation of at least 0.2 mm. Although a highly significant inverse correlation was observed between the amount of precipitation and PM concentrations of sizes 3–10 μm , 10–20 μm and 20–50 μm , the correlation coefficients were very weak (between −0.05 and +0.05); therefore, an interpretation should not be drawn from this analysis.

3.3. Fluctuations of Particle Matter Concentrations Following a Typical Heavy Rain Event

Granulometric analysis was used to explore how PM concentrations fluctuate following a typical heavy rain event. The PM concentration for each size was divided by the diameter of the particle. The result was then plotted (Figure 5) against the size of the particle, allowing us to compare PM concentration distributions of each of the 19 sizes.

Figure 5 compares the granulometric analyses 1 h before the rain event with 1 h after and with 15 h after. (The absence of a point on the orange line in Figure 5 is due to a measured value of zero). We can observe a 10-fold decrease in particle concentrations one hour after rain compared with one hour before for particle sizes 10–30 μm . It seems that precipitation reduced concentration levels of these larger particle sizes, yet this gap diminishes substantially 15 h after the rain event. Figure 6 demonstrates these changes quite clearly. The observed behaviors of PM concentrations relating to this single rain event seem to be confirmed in the results from the last part of our study that follows.

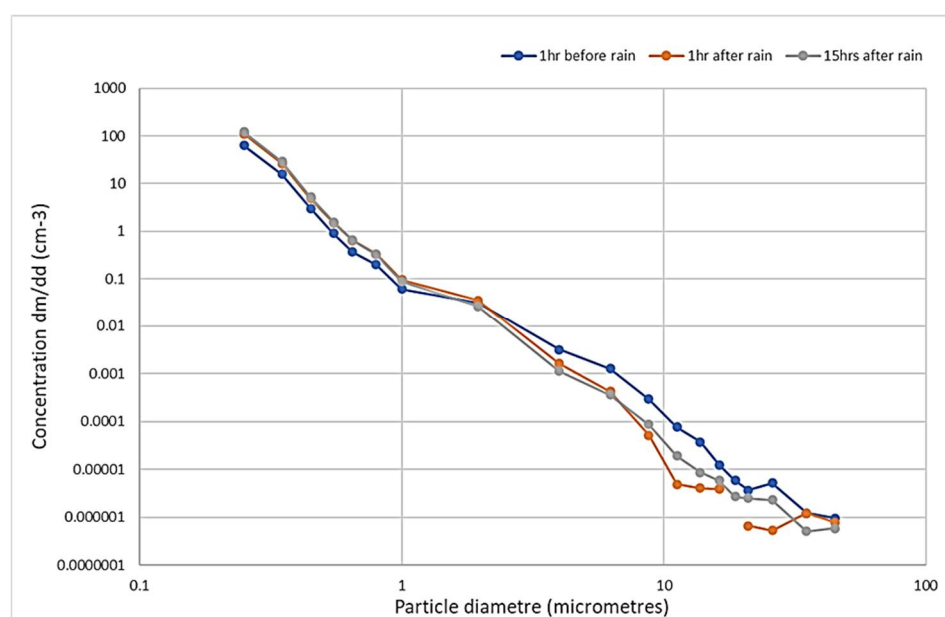


Figure 5. Granulometric analysis 1 h before the rain event, 1 h after and 15 h after.

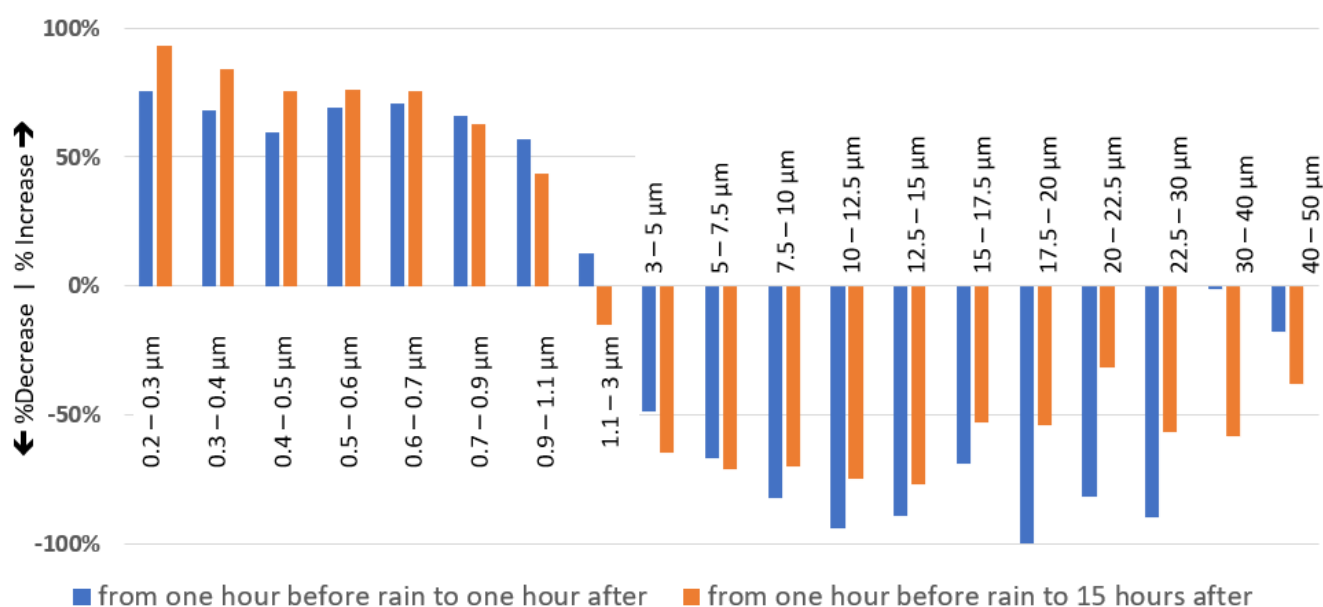


Figure 6. Percent change in PM concentrations by particle size range.

3.4. Recuperation of Particle Matter Levels after Rain

In this final analysis, we examine the progression of PM concentrations over time for 154 rain events, from 5 h before the beginning of precipitation to 20 h after the end of the precipitation. In Figure 7, line graphs plot the evolution of median PM concentrations hour by hour. The bars above and below the medians show the 40th and 60th percentiles. Each of the six graphs (one for each particle size range) is followed by a table showing the results of the Dunn's tests for the same size range by five-hour period.

For the three particle size ranges from 0.2 to 3 μm, we can observe in the graphs that median PM concentrations increased after rain and stayed high for the 20 h. The average of the PM concentration medians increased by 13% from the 5 h before rain to the 20 h after for particle sizes 0.2–0.5 μm, by 26% for particle sizes 0.5–1.1 μm and by 35% for particle sizes 1.1–3 μm. The associated tables also demonstrate this effect statistically—the Dunn's test results for particles of sizes 0.2–0.5 μm, comparing 5–1 h before rain with 1–20 h after rain, approach statistical significance, and the results for particles of sizes 0.5–3 μm are statistically significant.

For particles of sizes 3–10 μm, the average of the PM concentration medians increased by 17% from the 5 h before rain to the 5 h after but dropped back to pre-rain levels after 6 h. Indeed, concentration levels during rain are similar to levels 1–5 h after rain and are significantly higher than levels 6–20 h after rain.

PM concentrations of sizes 10–20 μm are on average 16.8% lower during the 10 h after rain than during the 5 h before rain. They then start going back up. There is a statistically significant increase (p -value = 0.006) in PM concentrations from the period 6–10 h after rain to 16–20 h after rain.

For particles of sizes 20–50 μm, there is a statistically significant difference between PM concentrations 5–1 h before rain and 6–15 h after rain.



Figure 7. Median, 40th and 60th percentile PM concentrations by hour before and after rain for 154 rain events and corresponding Dunn's test result tables for size ranges: (a) 0.2–0.5 µm; (b) 0.5–1.1 µm; (c) 1.1–3 µm; (d) 3–10 µm; (e) 10–20 µm and (f) 20–50 µm for the three locations between 2013 and 2019 (* indicates statistical significance).

4. Discussion

Our data collected between 2013 and 2019 revealed that the time during and the hours following precipitation are negatively correlated with PM concentrations in the air for particle sizes studied above 10 μm in diameter. A distribution comparative analysis (Dunn's test) of 154 events of 1 mm or more of rain confirmed a decrease in concentration levels for particles from 10 to 50 μm in diameter and allowed us to assess the recuperation of PM concentrations after rain. Concentrations of particles 10–20 μm in diameter returned to normal levels after approximately 10 h, while it took 15 h for particles 20–50 μm in diameter. Furthermore, granulometric analysis of a typical heavy rain event showed a 10-fold decrease only in concentrations of particles 10–30 μm in diameter one hour after rain compared with one hour before, and the reduction in PM concentrations appeared to last at least 15 h.

Conversely, no decrease was observed for particles smaller than 10 μm . PM concentrations of particles 0.2–10 μm actually increased following the precipitation events. PM concentrations of particles 0.2–3 μm stayed high for at least 20 h, while PM concentrations of particles 3–10 μm seemed to return to pre-rain levels after 6 h.

This study does not attempt to separate changes in PM concentrations due to micro-physical processes (washout) from large scale transport associated with air mass changes that may occur concurrently with precipitation processes. Therefore, it is not possible to conclude that the changes in PM are due to washout alone. Further statistical analyses could control for some confounding variables including wind speed, wind direction and factors associated with polluting activity, such as time of day and day of the week. Results might permit a more assertive assumption, yet the washout effect of precipitation seems a likely cause of the decreases observed following rain events.

The increase in measured concentrations of particles smaller than 10 μm after precipitation might be explained by the creation of water microdroplets as raindrops fall and break up. This phenomenon has been well documented [17]. Most rain droplets fall to the ground rapidly, yet smaller droplets fall at much slower speeds, remaining in the air for hours. The smaller the droplet, the slower the fall speed [18]. Like in fog, microdroplets of water tend to remain airborne [19]. Additionally, PM measurements using light scattering methods can be affected by humidity [20]. Indeed, high correlations were found between PM_{2.5} and PM₁₀ concentrations and humidity [21]. These various phenomena could explain why in our study the concentrations of smaller particle sizes were higher for hours after the rain—they might simply have consisted of water microdroplets left over from rain.

A previous study observed that PM_{2.5} concentrations were negatively correlated with the quantity of accumulated rainfall (R^2 value ranged from 0.668 to 0.974), identifying the function of the washing process of rain reducing PM_{2.5} pollution [10]. That analysis used the mass concentration of all particle sizes below 2.5 micrometers. The methodology of our study, however, examines the number of particles per cubic centimeter for different size ranges (e.g., 1.1–3 μm). Further, mass concentration measurements are more impacted by heavier particles, which may not be distributed equally across particle sizes. Comparisons between these two measurement methods are therefore problematic.

Fine particles and especially ultrafine particles are known to be more harmful than larger particles because they can penetrate deeper into the lungs and enter the bloodstream more easily [4]. Therefore, differences between large and small particles with regard to precipitation need further investigation. Follow-up studies should be conducted at different sites to confirm and add to the results of this study. Additionally, further studies could attempt to determine the composition of particle matter by size range before and after rain events. Finally, they could be conducted using our large sets of LOAC data to evaluate the below-cloud washout effect and to calculate its coefficient, as already conducted for the PM_{2.5} mass concentrations [22], this time considering all the various sizes of urban pollution particles, and controlling more for confounding variables.

5. Conclusions

After having considered exhaustive assessments of PM of different sizes and precipitation in three different locations between 2013 and 2019, we found that particles of sizes 10–50 μm diminished following precipitation events. This effect was measured to last locally for 10–15 h. Our study did not, however, observe that same effect for smaller particles.

The results of this study are useful to public health officials to advance their understanding of variations in air pollution by particle size correlate to weather conditions, helping them predict and prevent exposure of susceptible populations. The study also points to further research opportunities to be explored.

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Data Availability Statement: Precipitation data: Restrictions apply to the availability of these data. Data were obtained from “Météo-France” and are available at <https://donneespubliques.meteofrance.fr/> (accessed on 8 May 2020) with the permission of “Météo-France”.

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Conflicts of Interest: The authors declare no conflict of interest.

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